Simultaneous Measurement Method of Normal Spectral Emissivity and Optical Constants at High Temperatures[1]

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For spacecraft thermal design in the high temperature region, it is necessary to know accurate thermal radiative properties of heat-resistance materials at high temperatures. The emissivity depends on the surface conditions and the temperature of the materials. The purpose of the present study is to measure radiative properties, and to obtain the useful data indicated their surface conditions clearly. A simultaneous measurement method of the normal spectral emissivity using the Christiansen effect, the optical constants and the thickness of heat-resistance materials at high temperatures in the vacuum condition is proposed. The method is based on separated black body method and ellipsometry. This paper describes the measurement apparatus and preliminary results of measurement of molybdenum and zirconia specimen in the temperature range from 900K to 1400K and wavelength range from 2μm to 10μm. The experimental apparatus consists of a vacuum chamber, FT-IR with an optical system, an ellipsometryic system, a high temperature measurement system with the Christiansen effect, a heater control system, and a power supply.

KEY WORDS: normal spectral emissivity; optical constants; high temperature; ellipsometry; heat-resistance material; Christiansen effect; spacecraft thermal design

1. INTRODUCTION

It is the knowledge of thermal radiative properties of heat resisting materials that are important for thermal design of space devices used in high temperature surroundings.

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Especially, in thermal design for Re-usable Launch Vehicle (RLV), the emissivity of Thermal Protection System (TPS) materials is needed in order to estimate the surface temperature of the vehicle. For example of TPS, there are C/C composites, glass ceramics, Inconel 617, Titanium 1100 and so on [1-4]. The emissivity depends on the surface conditions and the temperature of the materials. One of the problems in measuring the emissivity is determining the surface temperature of the material accurately because large temperature gradients can develop in high-emissive and low-conductive materials. In this case, the paper proposes to use the new way the Christiansen effect. This effect is that the spectral emissivity is equal to unit at the Christiansen wavelength [5]. It is the purpose of the present study to measure the normal spectral emissivity of heat-resisting materials by separated blackbody method and to measure optical constants of their surface by ellipsometric method simultaneously for obtaining data indicated their surface conditions clearly. This paper describes the measurement apparatus and preliminary measurement results of normal spectral emissivity of molybdenum and zirconia specimen in the temperature range from 900K to 1400K. The results of optical constants of these materials at room temperature are also reported.

2. MEASUREMENTS

2.1. Principle of the Measurement

To measure spectral emissivity, the apparatus employs the separate blackbody technique, in which radiation from an area of a plane surface of a specimen, I_s, is compared with radiation from a similar area of a blackbody radiator, I_b, at the same wavelength λ and temperature T and under the same viewfactor. Thus, the emissivity ε is given by

$$\varepsilon(\lambda,\vartheta,T_s)=\frac{I_s(\lambda,\vartheta,T_s)-I_b(\lambda,T_{amb})}{I_b(\lambda,T_b)-I_b(\lambda,T_{amb})} \tag{1}$$

$$T_s = T_b \tag{2}$$

where $I_b(\lambda,T_b)$ is the intensity emitted by a blackbody at the temperature T_b, $I_b(\lambda,T_{amb})$ is the intensity emitted from the surrounding at ambient temperature, T_{amb}, $I_s(\lambda,\vartheta,T_s)$ is the intensity emitted from the specimen at the temperature T_s.

Optical constants are calculated by ellipsometric method [6]. The Fresnel

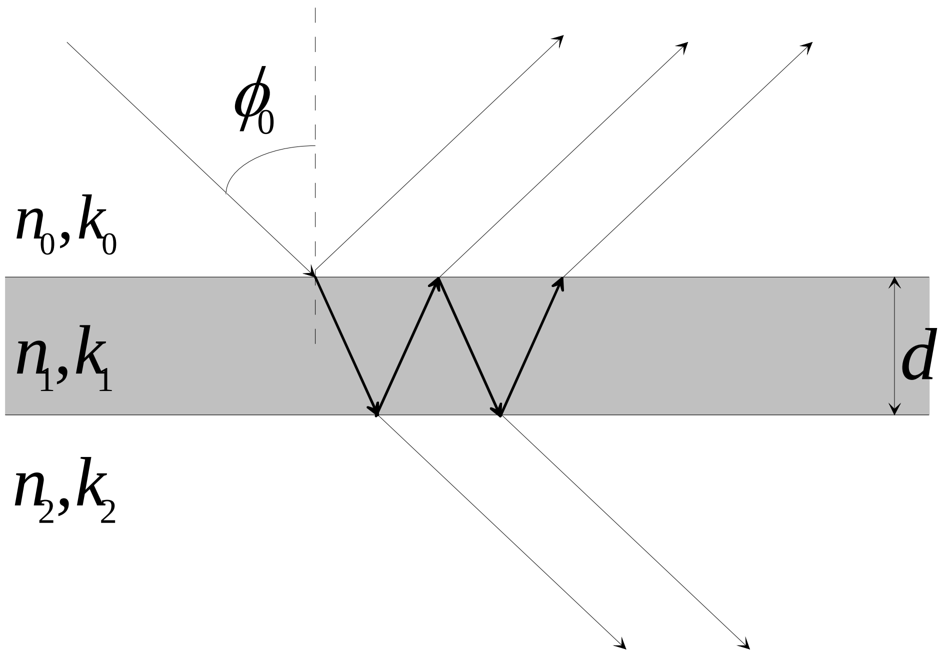

Figure 1. Incident beam to thin film.

reflection coefficients of a thin film covered surface (shown in Figure 1) are given by

$$R_p = \frac{r_{1p} + r_{2p}\exp(-i\delta)}{1 + r_{1p}r_{2p}\exp(-i\delta)} = r_p \exp(i\delta_p) \tag{3}$$

$$R_s = \frac{r_{1s} + r_{2s}\exp(-i\delta)}{1 + r_{1s}r_{2s}\exp(-i\delta)} = r_s \exp(i\delta_s) \tag{4}$$

where the subscripts p and s denote the components of the light, parallel (p) and perpendicular (s) to the plane of the incidence, respectively. The reflection coefficients for the boundary between i th and $i-1$th medium, r_{ip} and r_{ip}, are given by the Fresnel equations, i.e.,

$$r_{ip} = \frac{n_i \cos\phi_{i-1} - n_{i-1}\cos\phi_i}{n_i \cos\phi_{i-1} + n_{i-1}\cos\phi_i} \tag{5}$$

$$r_{is} = \frac{-n_i \cos\phi_i + n_{i-1}\cos\phi_{i-1}}{n_i \cos\phi_i + n_{i-1}\cos\phi_{i-1}} \tag{6}$$

$$n_{i-1}\sin\phi_{i-1} = n_i \sin\phi_i \tag{7}$$

$$\delta = \frac{4\pi n_i d\cos\phi_i}{\lambda} \tag{8}$$

$$i = 1,2 \tag{9}$$

where n_i and ϕ_i are the refractive index of the i th medium and the angle of incidence in that medium respectively. d is the thickness of the film, and λ is wavelength. Both R_p and R_s are not determinable because their phase shifts δ_p and δ_s cannot be measured, although the difference, $\Delta = \delta_p - \delta_s$, can be measured. Therefore, measurements are made of two parameters, Δ and ψ, defined by the ratio ρ_0 of R_p to R_s.

$$\rho_0 = \frac{R_p}{R_s} = \frac{r_p}{r_s} \exp i(\delta_p - \delta_s) = \tan\psi \exp(i\Delta) \tag{10}$$

Assuming that I_i and I_r are intensities of the incident and reflected light beams respectively gives the following equations.

$$I_r = r^2 I_i \tag{11}$$

$$r^2 = r_p^2 \sin\alpha + r_s^2 \cos\alpha \tag{12}$$

Therefore, for a small change of I_r due to the film,

$$\frac{\delta(r^2)}{r^2} = \frac{\delta I_r}{I_r}. \tag{13}$$

The polarizing prisms and the wave plate of the ellipsometer is set to proper orientations such that linearly polarized light is obtained after reflection from the specimen and the maximum intensity is registered. This is achieved by rotating one of the prisms $\pi/2$ radians from the extinction position used for the determination of Δ and ψ. By equations from (3) to (13), the optical constant and thickness of the film can be calculated.

2.2. Experimental Apparatus

The new experimental system developed to measure spectral emissivity and optical constants is presented in Figure 2. The main parts of this system consist of vacuum chamber with the specimen and the blackbody cavity, FTIR (Fourier transform infrared spectrometer), ellipsometer, radiation thermometer, pumping unit, heating unit, power supply and personal computers. The vacuum chamber shows Figure 3. The inner diameter is 255mm and the length is 512mm. The flange is in the shape of hemisphere with seven optical windows. The view angles of windows are 0˚, 30˚, 55˚ and 70˚. In the chamber the atmosphere is kept a vacuum of 10^{-4} Pa using a turbo-molecular pump.

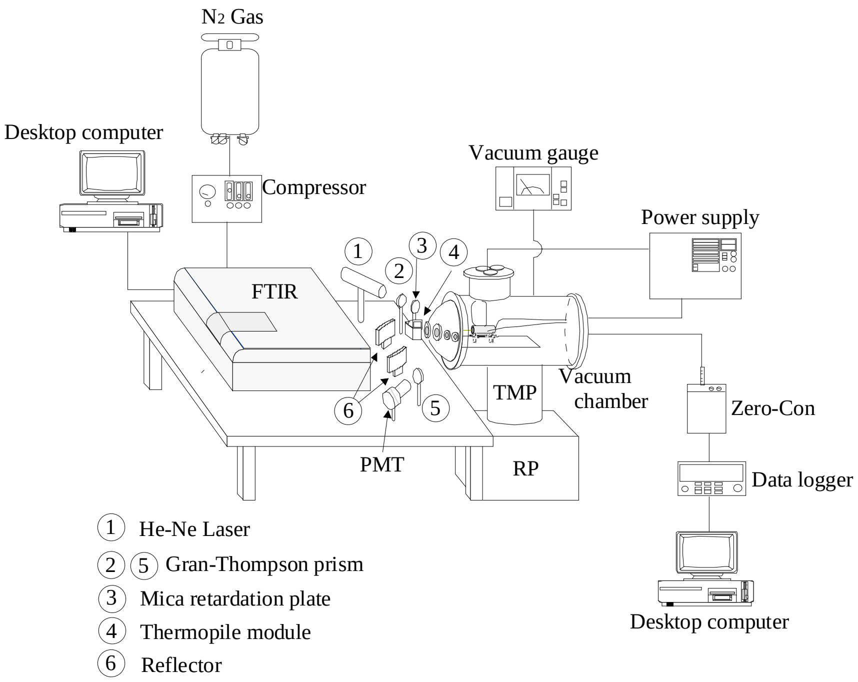

Figure 2. Measurement system.

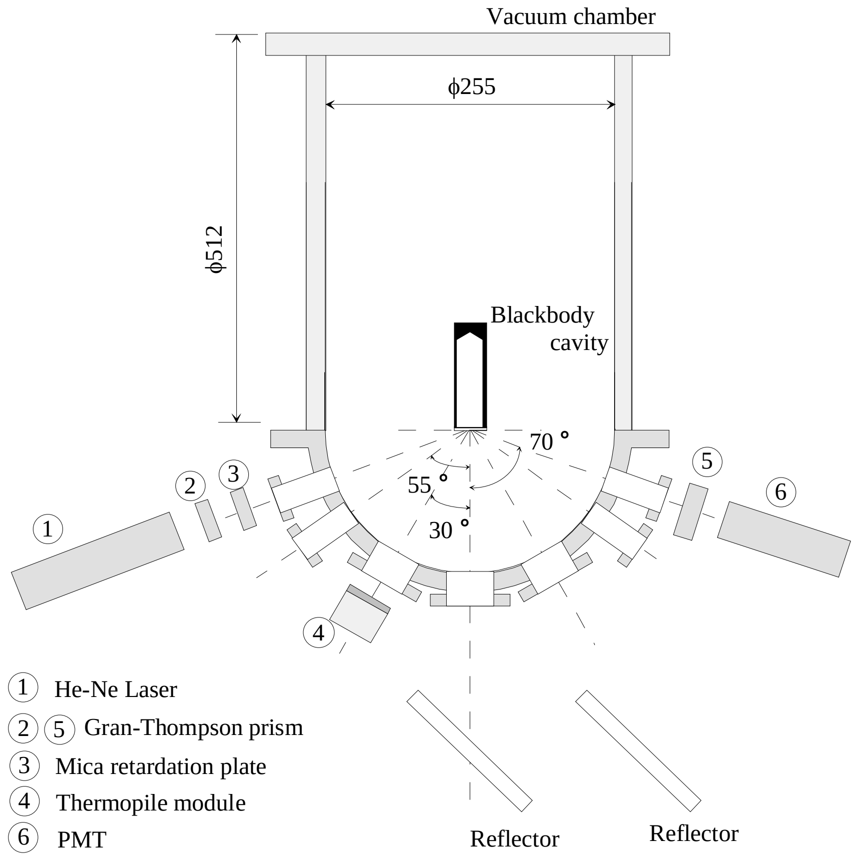

Figure 3. Cross section of the vacuum chamber.

The measurement of the normal spectral intensity is used FTIR spectrometer (Bio-Rad FTS-60A/896) in the wavelength range from 2μm to 10 μm. The radiation from the specimen or the blackbody is led through two reflectors to FTIR. The radiation is focused into the aperture stop and then onto the DTGS detector. The spectrum is measured on the average of 64 scans, and the wavelength resolution is $8cm^{-1}$. The layout of the optical system is showed in Figure 4. The diameter of the FTIR aperture and the point for measuring the emissivity is 10mm, and the diameter of the reflector is 130mm.

The ellipsometric system consists of He-Ne laser (05LHR691; MELLES GRIOT) whose wavelength is 632.8nm, two polarizers (Gran-Thompson Polarizing prisms), quarter-wave-plate (Mica retardation plate), and Photomultiplier Tube (R1104; Hamamatsu Photonics K.K.). The rotating polarizers and quarter-wave-plate is manually operated with accuracy of 5'. The incident angle of He-Ne laser is 55˚ or 70˚.

2.3. The Structure of Blackbody Cavity and Specimen

The heating unit is a cylindrical blackbody cavity as shown in Figure 5. The size is 140mm length, 28mm hole diameter, and 36mm outer diameter of the cylinder. The blackbody is made of molybdenum and alumina, and the inner walls of the cavity are painted by black paint (HiE-Coat 840-C; Aremco Products, Inc.) with more than 0.9 in emissivity. Two units of tantalum coil heater heat the blackbody. The temperature is measured by two Pt/Pt13%Rh (type R) thermocouples located at representative points

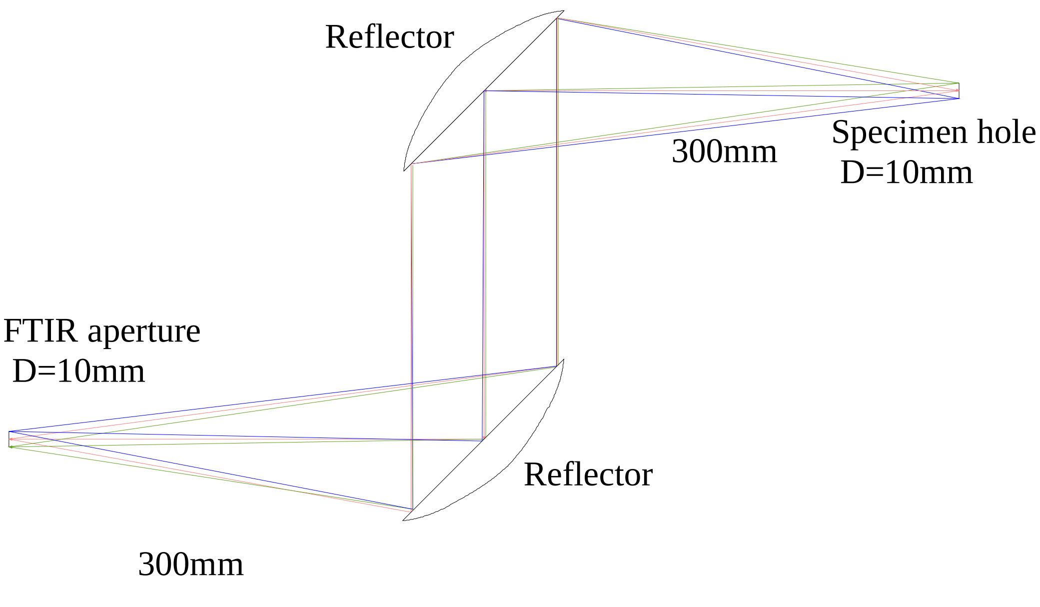

Figure 4. Layout of optical system.

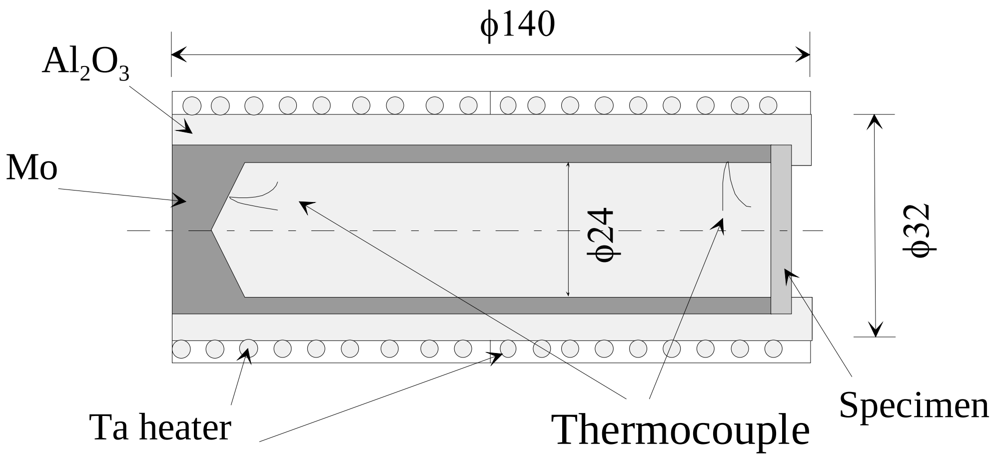

Figure 5. Heating unit (cross section).

on the inner walls, and controlled to realize a uniform temperature of ±3K.

The specimen is a plate of 28mm square and 4 mm thick. The position is set between molybdenum cavity and aluminum cylinder in front of blackbody cavity,. The specimen is heated by the radiation from the blackbody.

2.4. Temperature Measurement of Specimen

To determine the surface temperature of zirconia specimen by the Christiansen effect, the measurement system provides the thermopile module (A1TPMI; Perkin Elmer, Inc.) with narrow band pass filter. The thermopile module is located in front of the KRS5 window at the incident angle of 30˚. The wavelength range of The thermopile module is from 8μm to 14μm, and the narrow band pass filter has a peak wavelength of 11.96μm and half width is 0.97μm. Figure 6 shows the reflectivity of zirconia specimen in the wavelength region from 2μm to 16μm. The Christiansen wavelength is determined as 11.96μm because the reflectivity at this wavelength is less than 0.01 and the minimum. The radiation of blackbody cavity is measured with this module, and the relation between the temperature of blackbody cavity and the output of module as shown in Figure 7, is plotted. In the emissivity measurement the surface temperature of zirconia specimen is determined by Figure 7.

In the case of molybdenum specimen, the temperature is measured with a Pt/Pt13%Rh (type R) thermocouple within the hole (1.2mm diameter and 14mm depth) dug on the side of specimen.

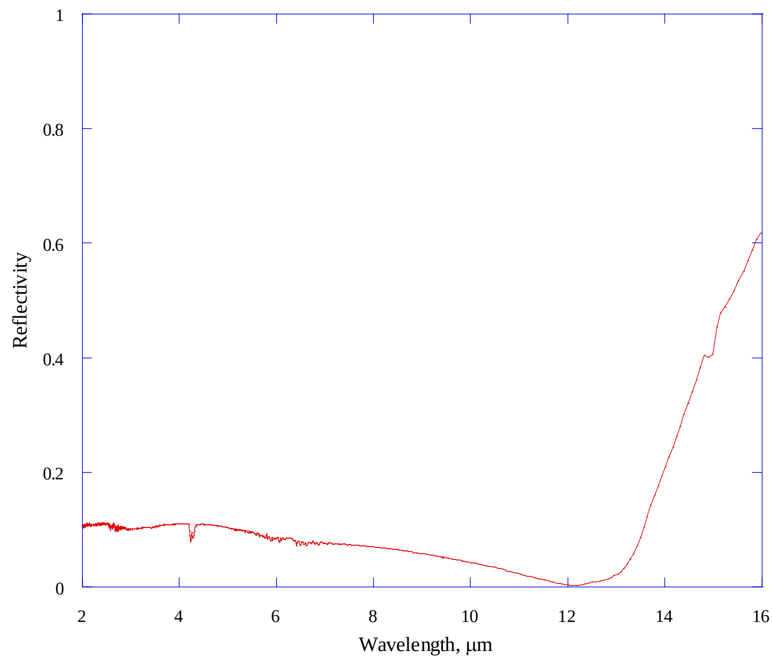

Figure 6. Reflectivity of zirconia.

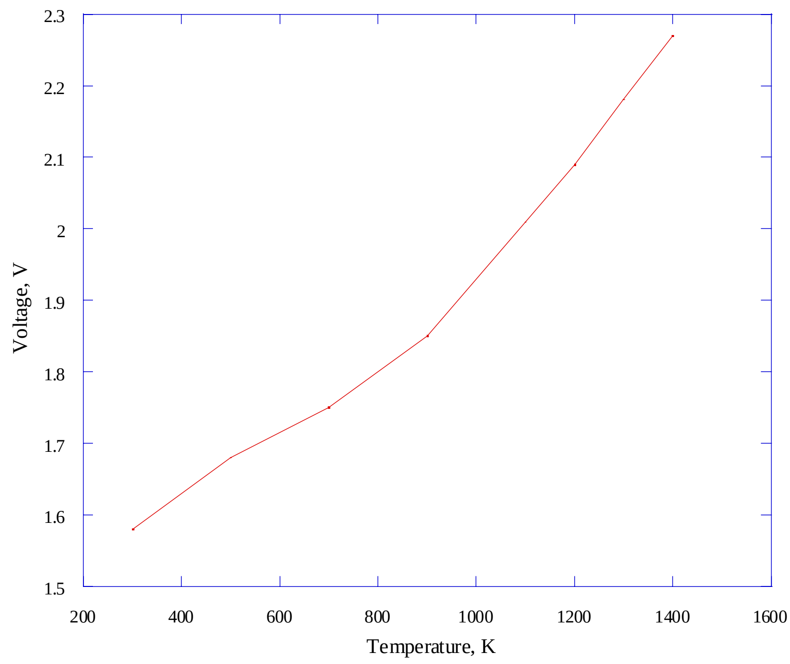

Figure 7. Temperature voltage of thermopile at Christiansen wavelength.

3. PRELIMINARY MEASUREMENTS AND RESULTS

3.1. Molybdenum Specimen

The normal spectral emissivity of the molybdenum specimen was measured at the wavelength region from 2μm to 10μm in the temperature of 900K, 1200K, and 1400K. Figure 8 shows the normal spectral emissivity of the molybdenum specimen. In the wavelength region from 2μm to about 9μm, the normal spectral emissivity tends to increase with the wavelength increasing. This tendency is inconsistent with the results of other reported data [7]. Approximately from 6μm to 7μm, the influence of absorption by carbon dioxide and vapor is observed. The results of measuring optical constants at room temperature before heating are $n = 2.83$ and $k = 2.45$ at 632.8nm. According to Reference [8], the optical constants of molybdenum are $n = 3.74$ and $k = 3.58$ at 652.6nm.

3.2. Zirconia Specimen

Figure 9 shows the normal spectral emissivity of zirconia specimen at the wavelength region from 2μm to 10μm in the temperature of 900K, 1200K, and 1400K. In the wavelength region from 2μm to about 9μm, the normal spectral emissivity tends to increase with the wavelength increasing. This tendency is different from that of

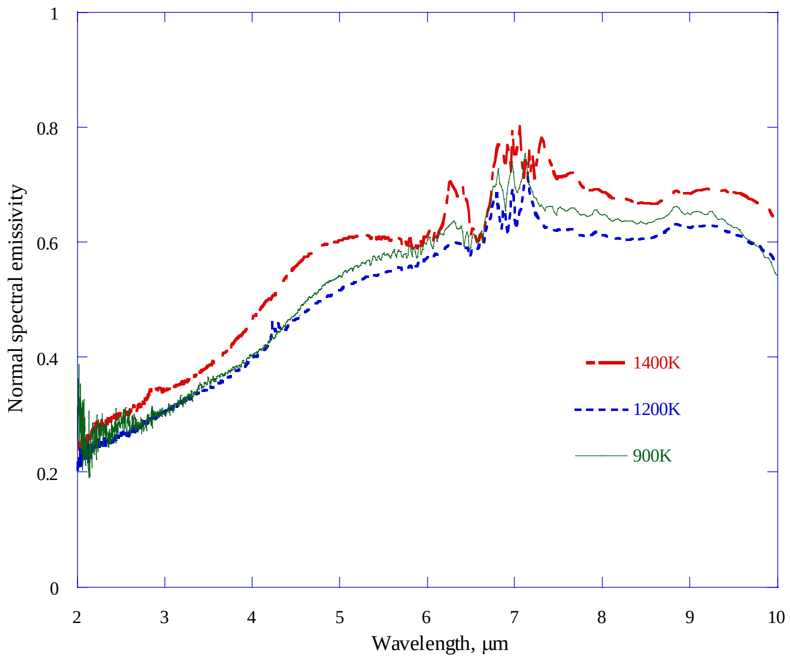

Figure 8. Normal spectral emissivity of molybdenum.

Figure 6, especially at the short wavelength. In Figure 6, the reflectivity is steady about 0.1 at the short wavelength. So, The emissivity is thought to be steady about 0.9 at the short wavelength. Approximately from 6µm to 7µm, the influence of absorption by carbon dioxide and vapor is observed. The results of measuring optical constants at room temperature before heating are $n = 1.74$ and $k = 0.24$ at 632.8nm.

4. CONCLUSION

A new apparatus has been developed for the simultaneous measurement of the normal spectral emissivity using the Christiansen effect, and of the optical constants at high temperatures. The normal spectral emissivity of molybdenum and zirconia specimen in the temperature from 900K to 1400K and the optical constants at room temperature were measured using this apparatus. From these results, the simultaneous measurement method of this apparatus is possibly effective. Future works are the establishment of the simultaneous measurement of emissivity and optical constants during heating, and of the method using the material with Christiansen effect as reference to determine sureface temperature of specimen.

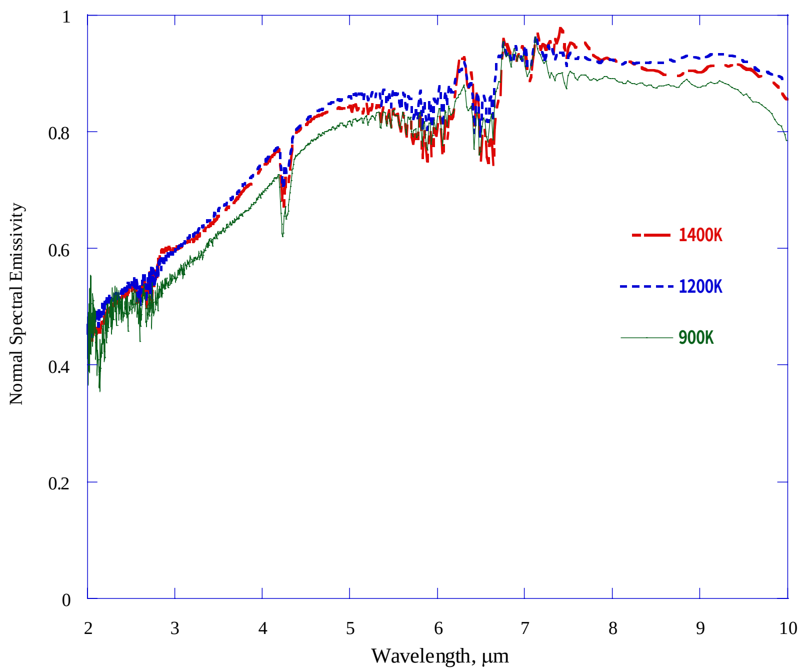

Figure 9. Normal spectral emissivity of zirconia

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